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PREPARATION AND PROPERTIES OF HIGH

Q MICROWAVE DIELECTRICS IN THE

(CA,SR,BA) ZIRCONATE SYSTEM

by

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ABSTRACT

Effects of powder synthesis and processing parameters on non-stoichiometric $CaZrO_3$ with added SrO and BaO were studied in order to optimize microstructure and microwave dielectric properties. Nitrate precursors of the constituent oxides were mixed in aqueous solutions to give the desired composition. The effect of solution pH (1-10) on powder characteristics and gel formation in the system were examined. Optimally high surface areas (31.4 m^2/g), small particle size (0.05μ), and uniform, dense microstructure were obtained only for the acidic (pH~1) powders. Ceramic parts with uniform microstructures, high dielectric constants ϵ_r=30-35, and high Q (>4000) were prepared from these powders.

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IN THE (CA, SR, BA) ZIRCONATE SYSTEM

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I. INTRODUCTION

Ceramic capacitor materials have become widely used as dielectric resonators in microwave circuit applications[1]. Advantages offered by the ceramic resonators are small size, ease of design and fabrication and mechanical ruggedness. Essential property requirements for the resonator components are a high dielectric constant ($\varepsilon_r > 25$), low dielectric loss (Q>3000) and small temperature coefficient of dielectric constant ($\tau_\varepsilon < 10$ ppm)[2]. These properties largely determine the size, resonance frequency, frequency stability, bandwidth and circuit performance of the resonator component. In consequence, a large number of ceramic systems have been investigated and microwave formulations developed to meet these property criteria[3-7].

The microwave dielectric properties of the $(Ca,Sr,Ba)ZrO_3$ system have been extensively investigated[2,4]. Data on relationships between the ceramic composition, phase field, dielectric properties and temperature coefficients are presented in Figure 1. Behavior of the relative dielectric constant, temperature coefficient and dielectric Q factor as a function of composition are detailed in Figures 1a, 1b and 1c respectively. The

Ca-rich region is shown to have a high dielectric Q (>2000) corresponding to the orthorhombic perovskite structure. The composition $(Ca_{0.965}Sr_{0.030}Ba_{0.005})Zr_{0.990}O_3$ was shown to possess the most desirable combination of high dielectric constant (29.3), low loss (Q>4000) and low temperature coefficient of permittivity (-22 ppm/°C) at 11 GHz[2]. This composition was nonstoichiometric with respect to ZrO_2 and exhibited a uniform, fine grained microstructure, although not optimally dense.

The dielectric quality factor, Q, has been shown to vary significantly between powder lots, even for ceramics with nearly identical microwave composition and microstructure[8]. This phenomenon has been traced to liquid forming impurities in the powder lots. These impurities can be reduced to by acid leaching,[8] but can be carefully controlled by processing steps relating to formulation, mixing, calcination and sintering[8]. Solution techniques offer advantages of close control of composition, high purity and ease of dispersing trace additives[9-11]. Excess solvent removal from the precursor salt solution is generally by precipitation-filtration or by evaporation of the solvent phase. For a multicomponent system, the resulting salt will nearly always be inhomogeneous, when dried by solvent evaporation, because of differential component solubilities at saturation. To minimize segregation and maintain homogeneity, the dried powder can be blended in a non-solvent medium to a stable paste consistency and dried. Spray drying may also be used to produce small droplets which dry rapidly, preserving a spherical shape for good powder handling[11]. Other powder characteristics such as crystallite size, surface area as well as agglomerate

size, shape, and hardness, are directly influenced by solution
pH. These parameters are interdependent and affect both densifi-
cation and microstructural development in the sintered cer-
amic[12,13].

Additionally, the calcination process is used to regulate
precursor decomposition, phase formation and powder reactivity,
factors which impact densification and grain growth dynamics.
Calcining just above the decomposition temperature of the precur-
sors, for example, typically gives the powder with highest reac-
tivity. Also, decomposition of the precursors (carbonates, sul-
fates, nitrates, citrates, chlorides, alkoxides) eliminates
gaseous by-products, evolution of which during sintering would
cause internal pressure, stresses and cracking.

Objective of this study was to investigate the influence of
processing parameters (powder synthesis, pH, calcination, and
sintering) on microstructural development in the widely used cer-
amic microwave resonator system, $(Ca,Sr,Ba)ZrO_3$.

II. EXPERIMENTAL

A nitrate solution technique was used to prepare the composi-
tion $Ca_{0.965}(Sr_{0.030}Ba_{0.005})Zr_{0.990}O_{3-x}$ (CSBZ), where
x=0.02. The nitrate precursors, consisting of $Ca(NO_3)_2 \cdot 4H_2O$,
$Sr(NO_3)_2$, $Ba(NO_3)_2$, and $Zr(NO_3)_2 \cdot nH_2O$, were separately
dissolved in deionized water and adjusted to pH $\approx$1.0 to facili-
tate dissolution. The individual solutions were assayed and
blended to give the desired composition. The mixed solution was
then converted to a powder by one of three different techniques.

The first involved evaporation of the solvent using a rotary eva-poration unit. This technique provided an acid powder with a high degree of homogeneity. The second method involved increas-ing the pH of the solution to an intermediate range (pH<5) by the addition of NH_4OH. For pH of ≈ 4.0 this resulted in a gelled structure that was then filtered and dried on a hot plate. The final technique involved coprecipitation from solution with the pH increased to $\geqslant 10.0$.

The dried powders were calcined at 600, 800 and 1000°C for 2 hours. The powders were further ball milled for a total period of 5 hours using ZrO_2 media in a solution of 60 vol% isopropyl alcohol and 40% deionized water, with Menhadden fish oil as a dispersant. A binder solution of 1 wt% PVA and 1 wt% Carbowax 4000 was added during the final hour of milling. The milled slurry was dried to ~3% moisture, passed through a 100 mesh screen and uniaxially pressed at 172 MPa. Pressed compacts were fired on ZrO_2 setters at temperatures between 1400 and 1550°C with soak times of 1.0 to 6.0 hours. Optimum solution pH, calci-nation temperature, firing temperature and soak time were arrived at by analysis of the fired densities and microstructures.

All powder samples were characterized using thermogravimet-ric, BET surface area, SEM (for morphology and particle size) and X-ray diffraction analysis. The sintered samples were character-ized as to density (helium pycnometry), X-ray diffraction phase analysis (using Cu K_α radiation at 6° 2θ/min) and by SEM analysis on as-fired, fractured and polished surfaces.

Capacitance and tanδ data were measured as a function of frequency from 1 MHz to 1 GHz using an RF impedance analyzer.

Samples were prepared by core drilling 0.7 cm disks, polishing
the surfaces, and sputtering with Au electrodes. High frequency
(12-16 GHz) measurements were carried out using the method given
by Howell[14]. A rectangular dielectric was metallized on all
sides, effectively forming a resonant cavity. This was then
inserted into a section of waveguide. Based upon the change in
transmission or reflection coefficients from the loaded to
unloaded condition, the dielectric properties could be deter-
mined.

III. RESULTS AND DISCUSSION

TGA analysis of the uncalcined powders showed the nitrates to
be completely decomposed at ~600°C. The minimum calcination
temperature of 600° was selected on this basis. Average crystal-
lite size, agglomerate size and surface area for unfired samples
calcined at 600°C for 2 hours and prepared from solution pH of
1.0, 2.0, 4.5, and 10.0 are summarized in Table I. Also, include-
d in Table I are data on corresponding sintered grain size and
density.

With increasing solution pH, average agglomerate size of the
powders increased and surface area correspondingly decreased.
Drying precursors from low pH solutions also tended to yield
softer and less agglomerated powders than the basic solutions, in
line with previous observations[12]. Generally, the powders from
solutions of intermediate pH were difficult to process as gella-
tion occurred (pH=4.5), giving powders which sintered poorly.
Crystallite size of these powders were anomalously large, indica-

tive of the associated state.

SEM photomicrographs depicting the morphology of powders prepared from solutions with different pH are shown in Figure 2 and confirm, in terms of agglomerate size, the trend depicted in Table I. As-fired surfaces of the samples are shown in Figure 3. The samples were calcined at 600°C for 2 hours and sintered at 1550°C for 2 hours. Grain size and microstructure for samples prepared from solutions of different pH were markedly different. Samples of intermediate pH showed poor densification but significant grain growth, while the acidic conditions (pH=1.0) offered the most uniform microstructure and highest density. Small particle size and high surface area associated with these powders provided the dynamics for superior densification. The lower density attained with the pH=10.0 powders can be attributed to the larger and harder agglomerates and lower surface area of the starting powders.

The calcination affect on average particle size, crystallite size, and surface area for unfired samples (prepared from a solution pH≈1.0) heat treated for 2 hours at 600, 800, and 1000°C are given in Table II. Figure 4 shows the corresponding x-ray diffraction patterns indicating the formation of orthorhombic phase at 600°C. Narrowing of the peaks at higher temperatures is indicative of the coarsening of particles. Data on the samples fired 1550°C/2h are presented in Table II. From these results it was determined that a calcination temperature of 600°C was optimal in terms of densification and microstructure uniformity. The powders calcined at 600°C had a larger surface area (hence a higher surface free energy) and would be more reactive. Table II

also shows the expected decrease in shrinkage that occurred with increased calcination temperature.

The effect of sintering temperature and time on densification were investigated for the range 1400-1550°C with a 2h soak time Sintering times were varied from 1-6 h at 1550°C. The results of these firings are summarized in Table III for the acid prepared powders calcined at 600°C for two hours as a function of sintering temperature and time. The grain growth shown by samples fired at intermediate temperatures is in agreement with the model proposed by Harmer et al.[15]. Densification process is predominate at high temperature while grain growth processes predominate at lower temperatures. Thus, samples fired at higher temperatures (>1500°C) had finer grain sizes because minimal time was spent in the temperature range dominated by grain growth. The unexpectedly high sintinering temperature for the chemically prepared powders can be attributed to the absence of any liquid phase constituents in the system.

SEM photomicrographs of as-fired surfaces as a function of sintering time are shown in Figure 5. Full densification was realized after 2 hours, but significant grain growth occurred throughout the firing cycle up to 4 hours as indicated. Average grain sizes determined were 2.5, 3.5, 12 and 16μm respectively for soak times of 1, 2, 4 and 6h at 1550°C. The orthorhombic phase was dominant by x-ray diffraction analysis, but traces of the cubic phase could be identified with longer soak times (>4h).

Figure 6 gives low frequency dielectric data for the optimally dense samples (soln pH=1; 600°C/2h calcine; 1550°C/2h

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sinter). Also included are dielectric data for the 12-16 GHz range. For the low frequency range, ε_r values were relatively constant except for a slight upturn above log frequency 8.3, near the operating stability limit for the bridge. Spurious resonances due to the cavity were also observed for Q, therefore, the data given represents a best line fit using a subroutine in the measurement package.

Dielectric data in the 12-16 GHz range was obtained using a [Hewlett Packard] network analyzer and rectangular samples with all but two corners metallized. This in effect constituted a parallel plate, rectangular loaded waveguide resonator. From these measurements the resonant modes and corresponding ε_r data were determined. The technique, however, is not well suited to accurate determination of high Q values, but Q data obtained exceeded 6000 for the dense samples. Because of this difficulty, microstructures could not be accurately correlated to the dielectric loss data, but the dense, pore free sample with smaller grain size appeared to give higher Q values. However, ε_r and Q values determined overall from the study were higher than previously reported [3], an improvement attributed to optimization of the processing conditions.

IV. CONCLUSIONS

1) In the $CaZrO_3$-BaO-SrO system, processing variables (solution pH, calcination, firing temperature and firing time) were found to significantly influence resultant microstruture.

2) Optimal properties were obtained with acidic solutions ($pH \approx 1.0$), low calcination temperature ($600^{\circ}C/2h$), and a firing temperature ($1550^{\circ}C/2h$).

3) Dielectric constant of ~35 for optimally dense samples was stable with frequency but Q decreased from ~5500 to ~4200 over the frequency range 1 MHz to 1 GHz. ε_r was also relatively stable in the 12-16 GHz range.

V. ACKNOWLEDGMENTS

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VII. List of Figures

Table I - Powder characteristics and sintered properties as a
function of pH for samples calcined at 600°C/2h and
sintered at 1550°C/2h.

	pH of Solution			
	1.0	2.0	4.5	10.0
Avg. Agglom. Size (μm):	0.05	0.07	0.12	0.50
Crystallite Size (Å):	68	104	213	72
Surface Area (m^2/g):	31.38	7.34	2.39	1.95
Avg. Grain Size (μm):	3.5	10.0	4.0	2.0
Sintered Density (%):	100	76.6	82.9	86.2

Table II - Powder characteristics and sintered properties
(1550°C/2h) for samples prepared from solution of
pH~1.0.

	Calcination Temperature		
	600°C	800°C	1000°C
Avg. Agglom. Size (μm):	0.05	0.16	0.18
Crystallite Size (Å):	68	103	206
Surface Area (m^2/g):	31.38	12.44	8.02
Avg. Grain Size (μm):	3.5	4.0	6.0
Sintered Density (%):	100	87.3	86.4
Diametral Shrink.(%):	25.0	19.2	16.6

Table III - Average grain size, density, for samples prepared
from a solution pH~1.0 and calcined at
$600^{o}C/2h$.

Sintering Temperature (^{o}C)	Average Grain Size (μm)	Density (%ThD)
1400	2.0	86.4
1450	7.0	93.0
1500	8.0	98.8
1550 (1h)	2.5	89.0
(2h)	3.5	100.0
(4h)	12.0	100.0
(6h)	16.0	100.0

a) Relative Dielectric
   Constant

b) Temperature Coefficient of
   Dielectric Constant (ppm/°C)

c) Dielectric Q

○ Ortho. s.s.,   ● Cubic s.s.,   ◐ Mixture.
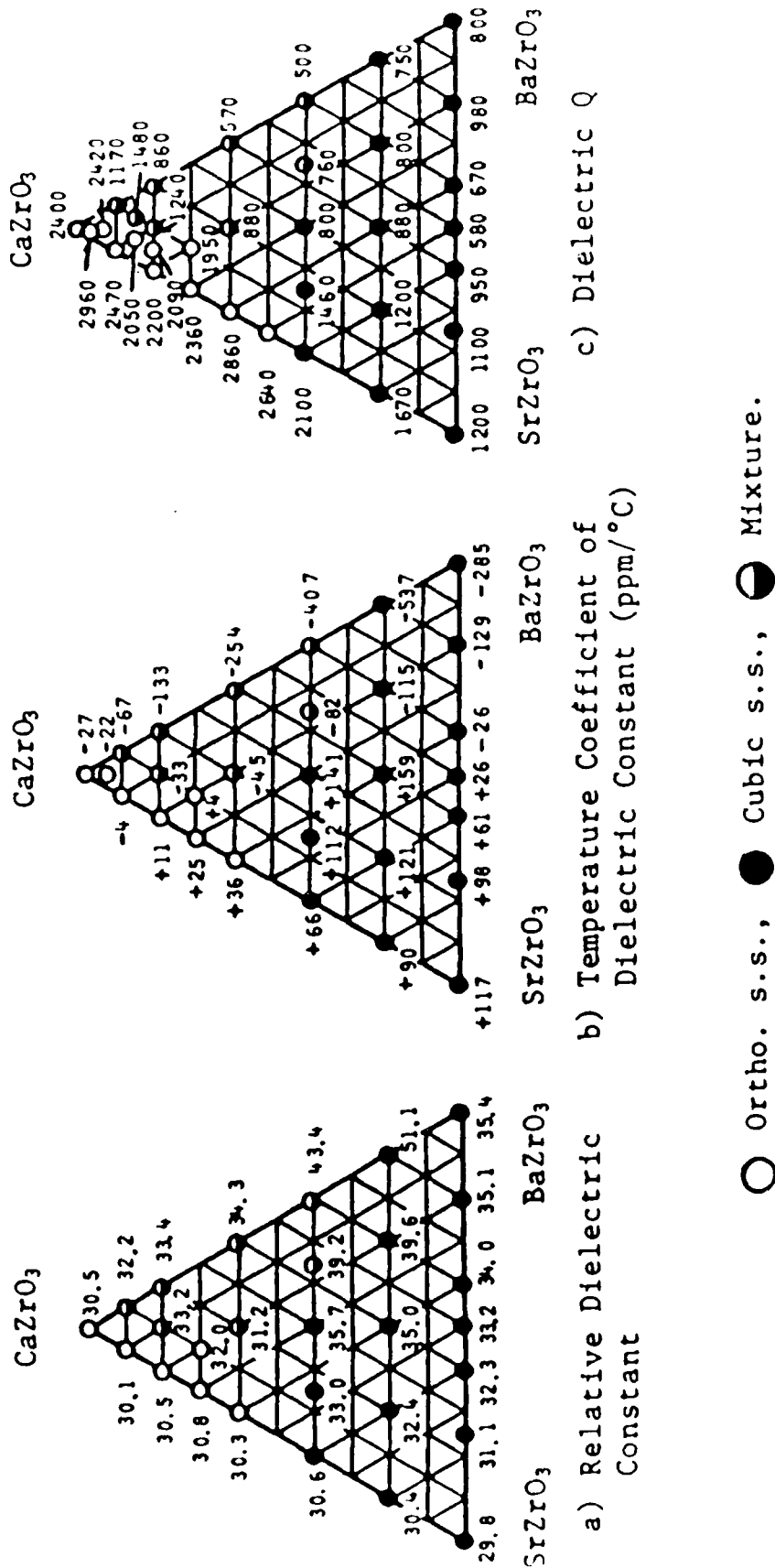
Figure 1. The relationship between composition and dielectric properties and crystal structure for the $(Ca,Sr,Ba) ZrO_3$ system at 11 GHz.
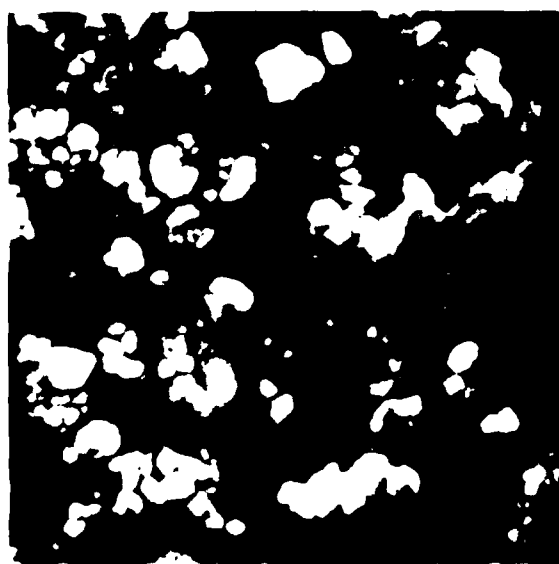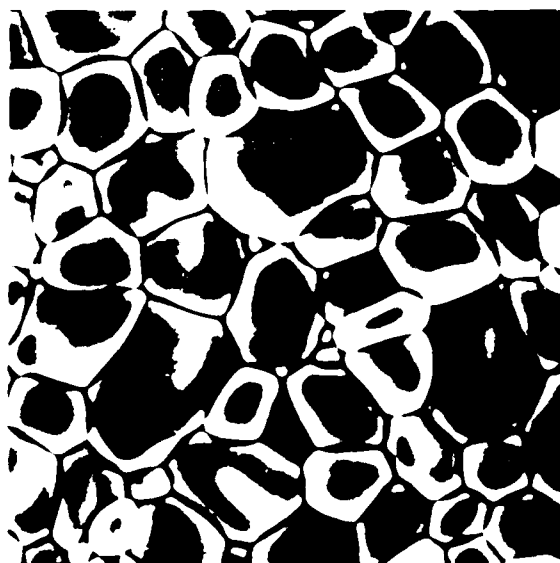
(a)   (b)

1.0 μm

(c)   (d)

Figure 2.   SEM photomicrographs of fracture surfaces of pressed
CSBZ samples calcined at 600°C - 2 hours and pre-
pared from a solution pH of: (a) 1.0; (b) 2.0; (c)
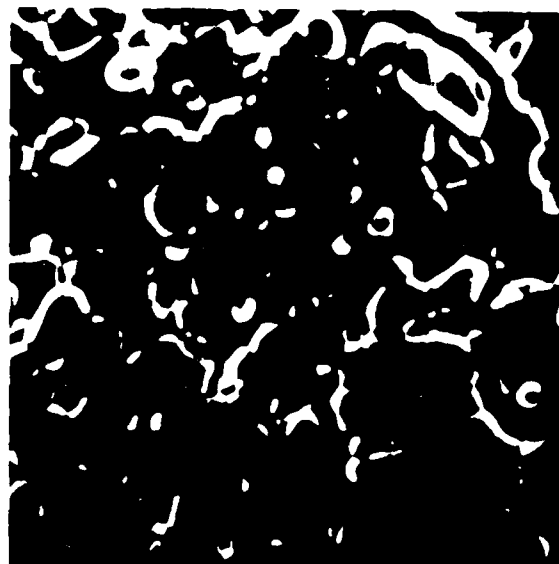4.5; and (d) 10.0, respectively.

(a)

(b)

10 µm

(c)

(d)

Figure 3.   SEM photomicrographs of as-fired surfaces of CSBZ
samples calcined at 600°C - 2 hours, fired at 1550°C
-2 hours, and prepared from a solution pH of: (a)
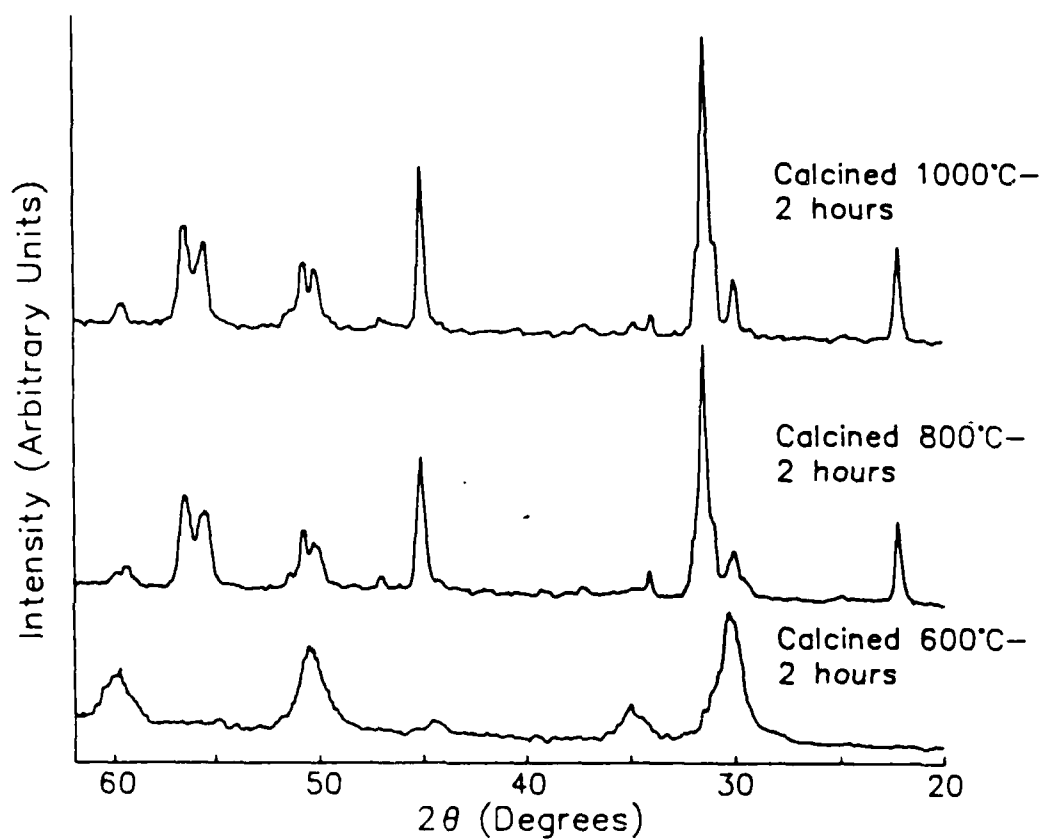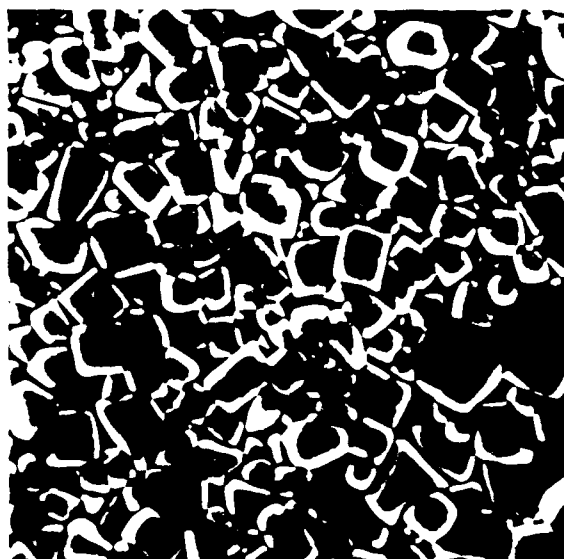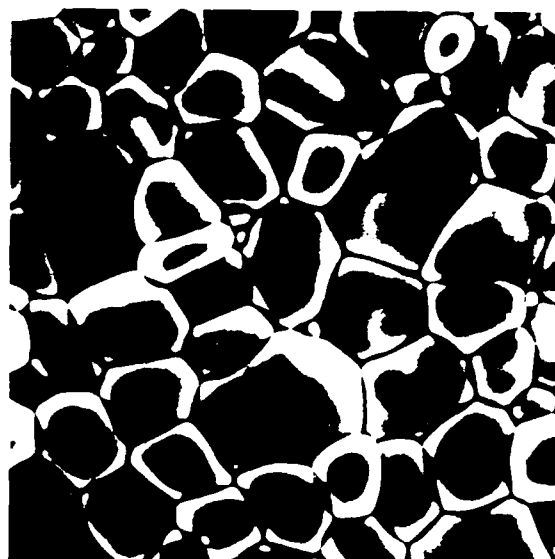1.0; (b) 2.0; (c) 4.5; and (d) 10.0, respectively.

Figure 4. X-ray diffraction patterns of CSBZ powders calcined at 600°C, 800°C, and 1000°C for 2 hours showing progressive development of the orthorhombic phase.

(a)

(b)

10 μm

(c)

Figure 5. SEM photomicrographs of as-fired surfaces of CSBZ
samples calcined at 600°C - 2 hours and fired at
1550°C for (a) 1 hour; (b) 2 hours; and (c) 4 hours
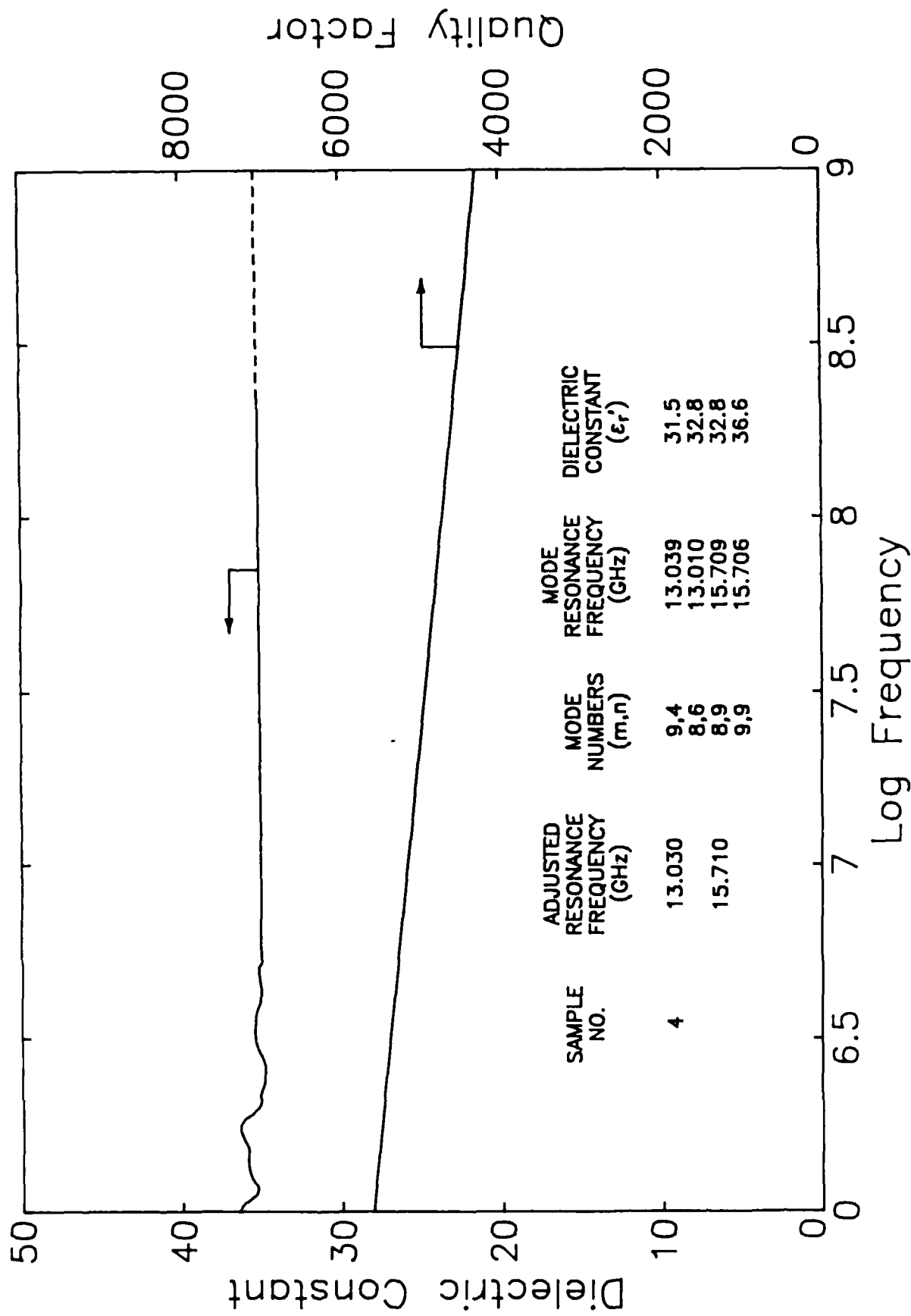respectively.

Figure 6. Dielectric properties versus log frequency for optimized CSBZ. Inset shows dielectric data in the 13-16 GHz range.

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